

## REMARKS

In accordance with the foregoing, the specification (pages 4-5) and the claim 1, 3-4 and 6 have been amended. Claim 2 is cancelled. Claims 8-11 have been added. Therefore, claims 1 and 3-11 are pending and under reconsideration, which is respectfully requested.

For the Amendment to the specification, it has been found that the term "diameter" appearing at page 4, line 15 is an error and should correctly read "radius". The above-mentioned term "diameter" (appearing at page 4, line 15 of the present specification) refers to a description of a prior art document, namely, Unexamined Japanese Patent Application Laid-Open Specification No. Sho 57-119837. **Support** for this amendment can be found at page 5, line 9 of the present specification (where there is the description reading: "average pore radius...").

As seen in the Amended claims above, in order to more clearly define the present invention, the feature (formula (1)) of claim 2 has been incorporated into claim 1, provided that **vanadium and niobium** have been deleted from the group of candidate component elements for the optional metal element D of the formula (1) which has been inserted into claim 2. By this amendment, **vanadium and niobium** have been excluded from claim 1.

No new matter has been added and accordingly, entry and approval of the claims 1 and 3-11 are respectfully requested.

## STATUS OF THE CLAIMS:

Claims 1, 3-11 are pending.

Claims 1-7 are rejected.

Claims 8-11 are added.

Claim 2 is cancelled.

## **II/ The state of the art and the features and advantages of the present invention:**

### **(I-1) Background information:**

Before specifically discussing the rejection of the claims, it is believed that the following background information should be considered in order to shed a proper light on the development of the present invention and the advantageous features thereof. As discussed in the specification under "Prior Art", it has been well known to produce acrylonitrile or

methacrylonitrile by ammonoxidation of propylene, isobutene or tert-butyl alcohol, namely, a reaction of propylene, isobutene or tert-butyl alcohol with molecular oxygen and ammonia. A number of suggestions have been made with respect to catalysts for use in the ammonoxidation of propylene, isobutene or tert-butyl alcohol. For example, various improvements have been provided with respect to ammonoxidation catalysts which are oxide catalysts containing molybdenum, bismuth and iron or containing antimony and iron.

A number of proposals have been made to improve an ammonoxidation catalyst by changing **the composition** thereof, and such proposals have contributed to the improvement of the catalyst performance. However, there have **not** been made many proposals to improve an ammonoxidation catalyst by changing **the physical structure** of the catalyst. As examples of prior art documents disclosing such proposals, there can be mentioned a prior art document (U.S. Patent No. 4,264,476, i.e., Umemura et al., cited in the office action) which discloses a catalyst for ammonoxidation of propylene, containing molybdenum, bismuth, iron, cobalt and zirconium as essential components, and a prior art document which discloses a catalyst for ammonoxidation of propylene, containing molybdenum, bismuth and antimony as essential components, wherein, in each of these catalysts, the amount of silica carrier, average pore diameter, total pore volume and specific surface area are, respectively, in specific ranges. Each of the above-mentioned two prior art documents (in which it is attempted to improve the catalyst performance by changing the physical structure of the catalyst) discloses a catalyst (having a specific physical structure) for use in producing acrylonitrile in a fluidized-bed reactor. However, there are no disclosures about the **pore distributions** of the catalysts in these two prior art documents.

Any of the conventional catalysts is still **unsatisfactory** in respect of the **yield** of the desired product. Therefore, it has been desired to develop a catalyst which can be advantageously used in the ammonoxidation of propylene, isobutene or tert-butyl alcohol in a **fluidized-bed reactor**, so that acrylonitrile or methacrylonitrile can be **stably** produced in **high yield**.

In this situation, the present inventors have made extensive and intensive studies with a view toward solving the above-mentioned problems of the prior art, specifically toward developing an excellent catalyst for use in the commercial production of acrylonitrile or methacrylonitrile by reacting propylene, isobutene or tert-butyl alcohol with molecular oxygen and ammonia in a fluidized-bed reactor, wherein the catalyst can produce acrylonitrile or methacrylonitrile stably in high yield. As a result, **it has unexpectedly been found that, in a**

particulate catalyst for use in the fluidized-bed ammonoxidation of propylene, isobutene or tert-butyl alcohol, when the cumulative pore volume of small-diameter pores having a pore diameter of 80 Å or less does not exceed a specific range, the catalyst exhibits a high yield of the desired product, and when the cumulative pore volume of large-diameter pores having a pore diameter of 1,000 Å or more does not exceed a specific range, the catalyst particles exhibit a high attrition resistance. Further, based on these findings, the present inventors have also found that, when **the pore distribution of a catalyst is controlled so as to satisfy specific requirements**, the catalyst exhibits **not only a highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst. Based on these findings, the present invention has been completed.

The instantly amended **claim 1** of the present application is directed to a particulate porous ammonoxidation catalyst for use in producing acrylonitrile or methacrylonitrile by reacting propylene, isobutene or tert-butyl alcohol with molecular oxygen and ammonia in a fluidized-bed reactor, the catalyst comprising a metal oxide and a silica carrier having supported thereon the metal oxide, wherein the metal oxide contains, as essential metal components, molybdenum, bismuth, iron and at least one element selected from the group consisting of sodium, potassium, rubidium and cesium (see formula (1) in claim 1), the catalyst having a particle diameter distribution wherein the amount of catalyst particles having a particle diameter of from 5 to 200  $\mu\text{m}$  is from 90 to 100 % by weight, based on the weight of the catalyst, and the catalyst having a **pore distribution** wherein the cumulative pore volume of pores having a pore diameter of 80 Å or less is not more than 20 %, based on the total pore volume of the catalyst and wherein the cumulative pore volume of pores having a pore diameter of 1,000 Å or more is not more than 20 %, based on the total pore volume of the catalyst.

**(I-2) Observations on the inventive step of the present invention:**

**(I-2-0)**

As described above, in the field of the production of acrylonitrile or methacrylonitrile by performing ammonoxidation, it has conventionally been needed to develop a catalyst which exhibits **not only a highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst **suitable for commercial use** as a fluidized-bed catalyst, and the present invention **fully meets the need**. In items (I-2-1) and (I-2-2) below, the respective specific meanings of "**high yield**" and "**high attrition resistance** which renders the catalyst **suitable for commercial use**", are described.

**(I-2-1)** With regard to the specific meaning of "high yield":

(economic importance)

It should be noted that it is **commercially very important** to achieve **even a very small improvement** (e.g., a 0.1 % improvement) in the **yield** of the desired product.

As well-known in the art, in the field of petrochemistry, an **enormous** amount of a raw material is used to produce an **enormous** amount of a product. Therefore, from the viewpoint of downsizing or streamlining of the production equipment and the viewpoint of achieving a reduction of the production cost, it is strongly desired that a catalyst exhibits a yield which is **as high as possible**. Especially in the case of the production of acrylonitrile or methacrylonitrile in a commercial scale plant (very large), **even a 0.1 %** improvement in yield will provide a production increase which is as large as **several hundreds of tons** per plant per year. Therefore, even a very small improvement in the catalyst performance, such as yield, can contribute **greatly** to the efficient use of petrochemical resources and the cost reduction of the production process. This point is described below in detail.

In an **ordinary plant** for acrylonitrile production in the U.S.A., the annual production amount of acrylonitrile is generally as large as about 100,000 tons to about 300,000 tons. In the case of a plant where the annual production amount of acrylonitrile is about 100,000 tons, when the yield of acrylonitrile is **84.0 %** (as in the case of **Comparative Example 3** of the present specification), the necessary amount of raw material **propylene** can be calculated as follows:

$$\text{Propylene} = 100,000 \text{ (tons/year)} / 53 / 0.84 \times 42 = 94,300 \text{ (tons/year)} \quad \text{formula (A)}$$

wherein 53 is the molecular weight of acrylonitrile, and 42 is the molecular weight of propylene.

On the other hand, when the yield of acrylonitrile is **84.2 %** (as in the case of **Example 2** of the present specification) (i.e., only a 0.2 % increase, relative to the above-mentioned 84.0 % yield), the necessary amount of raw material **propylene** can be calculated as follows:

$$\text{Propylene} = 100,000 \text{ (tons/year)} / 53 / 0.842 \times 42 = 94,100 \text{ (tons/year)} \quad \text{formula (B)}$$

wherein 53 and 44 are as defined in formula (A) above.

Thus, in the case of an ordinary plant for acrylonitrile production (where the annual production amount of acrylonitrile is 100,000 tons), by a **0.2 % improvement** in the yield of the

desired product, a reduction of about **200 tons/year** (i.e.,  $94,300 - 94,100 = 200$  (tons/year)) in the necessary amount of propylene can be achieved. This is quite a large value.

In this connection, it should be noted that, in view of the recent price level of propylene (i.e., 1,345 \$/ton), a reduction of **200 tons/year** in the necessary amount of propylene means a **cost reduction of \$269,000/year** (i.e.,  $1,345 (\$/ton) \times 200 (\text{tons/year})$ ). Such a great reduction in cost can be obtained at each plant. (It should be noted that a similar **cost reduction can also** be obtained with respect to another raw material, ammonia; however, for simplicity sake, the cost reduction obtained with ammonia is not considered here.)

Further, it should be noted that **Comparative Example 5** of the present specification exhibits a yield of acrylonitrile of 82.5 %, and **Example 4** of the present specification exhibits a yield of acrylonitrile of 84.5 %, that is, **Example 4** exhibits as a 2.0 % improvement in yield, relative to **Comparative Example 5**. In this case (i.e., when this 2.0 % improvement in yield is applied to the above-described calculation for determining a reduction in the necessary amount of raw material propylene), it is found that there can be obtained a **cost reduction of \$2,690,000/year** (i.e.,  $269,000 (\$/ton) \times 10$ ).

Needless to say, alternatively, the production of acrylonitrile can be greatly increased without increasing the amount of propylene used. In this case, by a **0.2 % improvement** in the yield of the desired product, an increase of **200 tons/year** in the amount of acrylonitrile produced can be achieved (see the calculation in the **Note<sup>1)</sup>** below). At the current U.S. domestic price level of acrylonitrile (i.e., \$1,908/ton), an increase of **200 tons/year** in the amount of acrylonitrile produced means a **sales increase of \$381,600/year** at each plant (i.e.,  $1,908 (\$/ton) \times 200 (\text{tons/year})$ ). Further, by a **2.0 % improvement** (i.e., 10 times the above-mentioned "0.2 %") in the yield of the desired product, the **sales increase becomes 10 times** the above-calculated value, that is, there can be obtained a **sales increase of \$3,816,000/year** at each plant.

**Note<sup>1)</sup>:** The increase of 200 tons/year in the amount of acrylonitrile produced can be calculated as follows. The amount of acrylonitrile produced from 94,300 (tons/year) of propylene in a yield of 84.0 % is **100,000** (tons/year) (as mentioned above; see formula (A)). The amount of acrylonitrile produced in a yield of **84.2 %** (i.e., a **0.2 % improvement**) is calculated by the following formula (C):

$$(94,300 \text{ (tons/year)}/42) \times 0.842 \times 53 = 100,200 \text{ (tons/year)}$$

formula (C)

(wherein 94,300 is the amount of propylene used, 42 is the molecular weight of propylene, and 53 is the molecular weight of acrylonitrile).

Then, the following subtraction is made:  $100,200 - 100,000 = 200$  (tons/year). Thus, it can be concluded that a **0.2 % improvement** in the yield of the desired product results in an increase of **200 tons/year** in the amount of acrylonitrile produced.

(environmental importance)

Further, it should be noted that a reduction in the amount of propylene used means a reduction in the occurrence of "greenhouse gas" ( $\text{CO}_2$ ) during the reaction for forming a desired product, thereby facilitating **the environmental protection**. That is, a reduction of about **200 tons/year** in the amount of propylene used means a **reduction** of about **630 tons/year** in the occurrence of  $\text{CO}_2$  (based on the assumption that all reduced amount of propylene is converted to  $\text{CO}_2$ ). The reduction of about 630 tons/year in the occurrence of  $\text{CO}_2$  is calculated by the following formula (D):

$$(200 \text{ (tons/year)}/42) \times 44 \times 3 = 630 \quad \text{formula (D)}$$

(wherein 42 is the molecular weight of propylene, 44 is the molecular weight of  $\text{CO}_2$ , and 3 is the number of  $\text{CO}_2$  molecules formed from one molecule of propylene).

Thus, even a small improvement in the yield of the desired product can achieve a **great commercial success** in the commercial production of (meth)acrylonitrile. This is becoming more evident in view of the recent tight condition of the oil market. Further, improvement of the yield of the desired product contributes greatly to the **environmental protection** by reducing the  $\text{CO}_2$  emissions.

As described hereinabove, it is strongly desired to achieve even a very small improvement (e.g., a 0.1 % improvement) in the yield of the desired product in the production of (meth) acrylonitrile.

**(I-2-2)** With regard to the specific meaning of "**high attrition resistance** which renders the catalyst **suitable for commercial use**":

Even when the initial yield of the desired product is high, if the catalyst loss during the reaction is too high, the cost of the catalyst becomes too high, thus rendering the catalyst disadvantageous economically. Such catalyst is unsuitable for commercial use.

Therefore, it is important that a catalyst exhibit satisfactorily high attrition resistance which renders the catalyst suitable for commercial use.

In the present invention, **judgment of whether or not** a catalyst exhibits **satisfactorily high attrition resistance** which renders the catalyst **suitable for commercial use**, is performed as follows. The measurement of the attrition resistance of the catalyst is performed in accordance with the method as described at page 30, line 3 to page 31, line 7 of the present specification, specifically by the method described in "Test Method for Synthetic Fluid Cracking Catalyst" (American Cyanamid Co., Ltd. 6/31-4m-1/57) (hereinafter referred simply as "ACC method"), which is generally known as a method for determining the attrition resistance of a fluidized-bed catalyst for use in the catalytic cracking. The attrition resistance of the catalyst of the present invention is measured in terms of the attrition loss which is defined by the following formula:

$$\text{attrition loss (\%)} = B / (C-A) \times 100$$

wherein:

A is the weight (g) of the catalyst which is lost by attrition in the period of from 0 hour point to 5 hour point after the start of the attrition resistance test;

B is, according to the ordinary ACC method, the weight (g) of the catalyst which is lost by attrition in the period of from 5 hour point to 20 hour point after the start of the attrition resistance test; however, in the present invention, this B is defined as the weight (g) of the catalyst which is lost by attrition in the period of from 5 hour point to 120 hour point after the start of the attrition resistance test; and

C is the weight (g) of the catalyst used in the attrition resistance test.

When the catalyst exhibits an attrition loss of 7 % or less, it is judged that the catalyst has a **satisfactorily high attrition resistance** which renders the catalyst suitable for commercial use. This criterion has been established based on the long time experience of the Applicant as a skilled person in the art. (The above-mentioned value: "attrition loss of 7 % or

**less**" is hereinafter frequently referred to as the "commercially suitable, reduced attrition loss of **7 % or less**")

For example, the catalysts obtained in **Examples 1 to 4** of the present specification (see Table 2 at page 73) exhibit an attrition loss in the range **as low as from 3.5 to 5.7 %** (which does not exceed the above-mentioned "commercially suitable, reduced attrition loss of **7 % or less**"). Such catalysts of **Examples 1 to 4** of the present specification are judged to exhibit an **excellent attrition resistance** (and are suitable for commercial use). On the other hand, the catalyst obtained in **Comparative Example 3** of the present specification (see Table 2 at page 73) exhibits an attrition loss **as high as 15 %** (which exceeds the above-mentioned "commercially suitable, reduced attrition loss of **7 % or less**"). Such catalyst of **Comparative Example 3** of the present specification is judged to exhibit a **poor attrition resistance** (and is unsuitable for commercial use).

**(I-2-3)** With respect to the critical importance of the pore distribution requirement of claim 1:

Claim 1 has the pore distribution requirement that the catalyst have a pore distribution wherein the **cumulative pore volume of pores having a pore diameter of 80 Å or less is not more than 20 %**, based on the total pore volume of the catalyst, and the **cumulative pore volume of pores having a pore diameter of 1,000 Å or more is not more than 20 %**, based on the total pore volume of the catalyst. With respect to the importance of this pore distribution requirement, the present specification has the following description:

"When the cumulative pore volume of pores having a pore diameter of 80 Å or less is more than 20 %, based on the total pore volume of the catalyst, the catalyst poses a problem in that the yield of the desired product (i.e., acrylonitrile or methacrylonitrile) becomes low. On the other hand, when the cumulative pore volume of pores having a pore diameter of 1,000 Å or more is more than 20 %, based on the total pore volume of the catalyst, a problem arises in that the attrition resistance of the catalyst becomes poor and, hence, the catalyst cannot exhibit a satisfactorily high attrition resistance which renders the catalyst suitable for commercial use, so that it becomes impossible to stably produce the desired product. The reason why the yield of the desired product becomes low when the

cumulative pore volume of pores having a pore diameter of 80 Å or less is more than 20 %, based on the total pore volume of the catalyst, has not yet been elucidated. However, it is presumed that, in the small-diameter pores having a pore diameter of 80 Å or less, the residence time of the reaction product in the pores of the catalyst becomes too long, thus accelerating decomposition of the reaction product, leading to a lowering of the yield of the desired product."

*(emphasis added)(see page 23, line 18 to page 24, line 16 of the present specification)*

#### **(I-2-4)**

As described below in more detail with reference to the Examples and Comparative Examples of the present specification, the ammonoxidation catalyst of the present invention exhibits not only a **high yield** of the desired product, but also a **high attrition resistance** which renders the catalyst suitable for commercial use. Therefore, the catalyst of the present invention is advantageous in that, when the catalyst is used for performing a catalytic ammonoxidation of propylene, isobutene or tert-butyl alcohol in a fluidized-bed reactor, acrylonitrile or methacrylonitrile can be produced **stably in high yield**.

#### **(I-3)** Data showing the **excellent effects** of the present invention:

The Applicant wishes to contend that the requirements of **claim 1** of the present application (especially the **pore distribution requirement**) are **critical** for achieving the excellent effects of the present invention, i.e., the effects that the catalyst exhibits **not only a highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst. In order to substantiate this contention of the Applicant, the Applicant wishes to hereinafter make observations, with reference to Examples 1 to 4 and Comparative Examples 1 to 5 of the present specification, to show that the requirements of **claim 1** of the present application (especially the **pore distribution requirement**) are **critical** for achieving the excellent effects of the present invention (i.e., the effects that the catalyst exhibits **not only a highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst).

In each of **Examples 1 to 4** and **Comparative Examples 1 to 5** of the present specification, an ammonoxidation catalyst for use in a fluidized-bed reactor is produced (see page 52, line 18 to page 71, line 6 of the present specification). The catalyst produced is subjected to an attrition resistance test (120 hours) in accordance with the ACC method (as described at page 30, line 3 to page 31, line 3 of the present specification). Also, by using the catalyst produced, an ammonoxidation reaction of propylene is performed.

With respect to each of the catalysts produced in Examples 1 to 4 and Comparative Examples 1 to 5, the composition thereof and the production conditions therefor are shown in Table 1 (at page 72 of the present specification). The pore distribution, and the catalyst performance and strength (i.e., yield of desired product, conversion of raw material, and attrition loss) are shown in Table 2 (at page 73 of the present specification).

For easier reference, Tables 1 and 2 of the present specification are reproduced below.

Table 1

	Mo	Bi	Fe	C	D	E	G	Amount of silica carrier present in the catalyst (% by weight)	Average primary silica particle diameter of silica sol and the amount of silica sol (% by weight)	Final calcination temperature (°C)
Ex. 1	12	0.45	1.8	$\text{Ni}_2\text{CO}_3\text{Mg}_2$	—	$\text{Ce}_{0.9}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	22 nm = 100	580
Compara.	12	0.45	1.8	$\text{Ni}_2\text{CO}_3\text{Mg}_2$	—	$\text{Ce}_{0.9}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	12 nm = 100	590
Ex. 1	12	0.45	1.8	$\text{Ni}_2\text{CO}_3\text{Mg}_2$	—	$\text{Ce}_{0.9}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	86 nm = 100	550
Compara.	12	0.45	1.8	$\text{Ni}_2\text{CO}_3\text{Mg}_2$	—	$\text{Ce}_{0.9}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	86 nm:12 nm = 50:50 <sup>1)</sup>	590
Ex. 2	12	0.6	1.8	$\text{Ni}_5\text{Mg}_2$	—	$\text{Ce}_{0.75}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	86 nm:12 nm = 75:25 <sup>2)</sup>	570
Compara.	12	0.6	1.8	$\text{Ni}_5\text{Mg}_2$	—	$\text{Ce}_{0.75}$	$\text{K}_{0.09}\text{Rb}_{0.05}$	50	86 nm:12 nm = 50:50 <sup>3)</sup>	580
Ex. 3	12	0.3	2	$\text{Ni}_{5.4}\text{Zn}_{2.1}$	—	$\text{Pr}_{0.13}\text{Nd}_{0.47}$	$\text{K}_{0.08}\text{Cs}_{0.04}$	50	58 nm:8 nm = 50:50 <sup>3)</sup>	610
Compara.	12	0.3	2	$\text{Ni}_{5.4}\text{Zn}_{2.1}$	—	$\text{Pr}_{0.13}\text{Nd}_{0.47}$	$\text{K}_{0.08}\text{Cs}_{0.04}$	50	8 nm = 100	560
Ex. 4	12	0.3	1.2	$\text{Ni}_{6.2}\text{Mg}_{2.5}$	$\text{Cr}_{0.2}\text{In}_{0.2}$	$\text{Ce}_{0.3}$	$\text{K}_{0.2}$	35	12 nm = 100	600
Compara.	12	0.3	1.2	$\text{Ni}_{6.2}\text{Mg}_{2.5}$	$\text{Cr}_{0.2}\text{In}_{0.2}$	$\text{Ce}_{0.3}$	$\text{K}_{0.2}$	50	12 nm = 100	600
Ex. 5										

Note:

- 1)  $\text{SiO}_2$  having an average primary silica particle diameter of 86 nm :  $\text{SiO}_2$  having an average primary silica particle diameter of 12 nm = 50 % by weight : 50 % by weight
- 2)  $\text{SiO}_2$  having an average primary silica particle diameter of 86 nm :  $\text{SiO}_2$  having an average primary silica particle diameter of 12 nm = 75 % by weight : 25 % by weight
- 3)  $\text{SiO}_2$  having an average primary silica particle diameter of 58 nm :  $\text{SiO}_2$  having an average primary silica particle diameter of 8 nm = 50 % by weight : 50 % by weight

Table 2

Pore volume (cc/g)	The ratios of the cumulative pore volume of pores having a pore diameter of 80 Å or less and the cumulative pore volume of pores having a pore diameter of 1,000 Å or more, based on the total pore volume of the catalyst			Reaction results			Results of the attrition resistance test		
	Pores having a pore diameter of 80 Å or less	Total pore volume of the catalyst	Pores having a pore diameter of 80 Å or less	Pores having a pore diameter of 1,000 Å or more	Conversion	Yield of AN			
Ex. 1	0.005	0.012	0.232	2.2 %	5.2 %	4.3	99.0 %	84.0 %	5.7 %
Compar. Ex. 1	0.058	0.001	0.22	26.4 %	0.5 %	3.7	99.1 %	82.4 %	2.9 %
Compar. Ex. 2	0	0.271	0.354	0.0 %	76.6 %	3.9	99.1 %	84.5 %	7 % (5 - 20 Hr.)
Ex. 2	0.014	0.004	0.225	6.2 %	1.8 %	4.6	99.2 %	84.2 %	5.2 %
Compar. Ex. 3	0.002	0.152	0.289	0.7 %	52.6 %	4.2	99.2 %	84.0 %	15.0 %
Ex. 3	0.022	0.009	0.237	9.3 %	3.8 %	4.1	99.1 %	84.2 %	4.8 %
Compar. Ex. 4	0.0802	0	0.204	39.3 %	0.0 %	3.8	99.1 %	81.9 %	1.5 %
Ex. 4	0.02	0.013	0.21	9.5 %	6.2 %	4.2	99.1 %	84.5 %	3.5 %
Compar. Ex. 5	0.061	0.001	0.213	28.6 %	0.5 %	3.9	99.0 %	82.5 %	1.9 %

As shown in Table 1 above, all of the catalysts produced in **Examples 1 to 4** and **Comparative Examples 1 to 5** satisfy the below-mentioned two of the requirements of claim 1 of the present application:

the **metal element composition requirement** that the metal oxide contain at least two elements selected from the group consisting of molybdenum (Mo), bismuth (Bi), iron (Fe), vanadium (V), tellurium (Te) and niobium (Ni), and

the **silica carrier content requirement** that the silica carrier be present in an amount of from 20 to 80 % by weight, based on the total weight of the metal oxide and the silica carrier.

Further, as described at page 50, lines 5 to 8 of the present specification, with respect to all of the catalysts produced in **Examples 1 to 4** and **Comparative Examples 1 to 5**, the amount of catalyst particles having a **particle diameter of from 5 to 200 µm is 100 % by weight**, based on the weight of the catalyst. Therefore, all of the catalysts produced in **Examples 1 to 4** and **Comparative Examples 1 to 5** satisfy the particle diameter distribution requirement that the amount of catalyst particles having a particle diameter of from 5 to 200 µm be from 90 to 100 % by weight, based on the weight of the catalyst.

On the other hand, with respect to the **pore distribution** of each of the catalysts produced in **Examples 1 to 4** and **Comparative Examples 1 to 5**, the following should be noted. For easier reference, the most important data of Table 2 above are collected and summarized in Table A below.

Table A

	The ratios of the cumulative pore volume of pores having a pore diameter of 80 Å or less and the cumulative pore volume of pores having a pore diameter of 1,000 Å or more, based on the total pore volume of the catalyst		Reaction results		Results of the attrition resistance test
	Pores having a pore diameter of 80 Å or less	Pores having a pore diameter of 1,000 Å or more	Conversion	Yield of AN	Attrition loss
Ex. 1	2.2 %	5.2 %	99.0 %	84.0 %	5.7 %
Ex. 2	6.2 %	1.8 %	99.2 %	84.2 %	5.2 %
Ex. 3	9.3 %	3.8 %	99.1 %	84.2 %	4.8 %
Ex. 4	9.5 %	6.2 %	99.1 %	84.5 %	3.5 %
Compara. Ex. 1	<b>26.4</b> %	0.5 %	99.1 %	82.4 %	2.9 %
Compara. Ex. 2	0.0 %	<b>76.6</b> %	99.1 %	84.5 %	7 % (5 - 20 Hr)
Compara. Ex. 3	0.7 %	<b>52.6</b> %	99.2 %	84.0 %	15.0 %
Compara. Ex. 4	<b>39.3</b> %	0.0 %	99.1 %	81.9 %	1.5 %
Compara. Ex. 5	<b>28.6</b> %	0.5 %	99.0 %	82.5 %	1.9 %

Note: With respect to Comparative Examples 1 to 5, the values in ***bold italics*** are those values which do not satisfy the pore distribution requirement of claim 1.

As shown in (Table 2 and) Table A above, all **Examples 1 to 4** satisfy the pore distribution requirement (of claim 1 of the present application) that the catalyst have a pore distribution wherein the cumulative pore volume of pores having a pore diameter of 80 Å or less is not more than 20 %, based on the total pore volume of the catalyst and wherein the cumulative pore volume of pores having a pore diameter of 1,000 Å or more is not more than 20 %, based on the total pore volume of the catalyst.

Thus, all **Examples 1 to 4** satisfy all requirements of claim 1 of the present application.

On the other hand, as also shown in (Table 2 and) Table A above, any of **Comparative Examples 1 to 5** does not satisfy the pore distribution requirement of claim 1 of the present application.

(**performance and strength** of the catalysts)

The performance and strength of such catalysts of **Examples 1 to 4** and **Comparative Examples 1 to 5** are as follows.

In the case of the catalysts of **Examples 1 to 4**, the **yield** of the desired product (acrylonitrile) is in the range of from 84.0 to 84.5 %, and the **attrition loss** is in the range of from 3.5 to 5.7 %. As described in item (II-2-2) above, when the catalyst exhibits an **attrition loss of 7 % or less**, the catalyst has a satisfactorily high attrition resistance which renders the catalyst suitable for commercial use.

By contrast, in the case of the catalysts of **Comparative Examples 1 to 5**, the **yields** of the desired product (acrylonitrile) are, respectively, 82.4 %, 84.5 %, 84.0 %, 81.9 % and 82.5 %. That is, the **yields** (84.5 % and 84.0 %) exhibited, respectively, in

**Comparative Examples 2 and 3** are at the **same** level as in the case (in the range of from 84.0 to 84.5 %) of **Examples 1 to 4**. **Seemingly** good results are obtained in **Comparative Examples 2 and 3**. However, in connection with such seemingly good results of **Comparative Examples 2 and 3**, it should be noted:

that the catalyst of **Comparative Example 2** exhibits an **attrition loss as large as 7 %** in the 5 - 20 hour period (i.e., early period) of the attrition resistance test; this means that, if the test is performed for as long as 120 hours as in the case of the Examples and the other Comparative Examples, the **attrition loss** would be far higher than 7 %; and

that the catalyst of **Comparative Example 3** exhibits an **attrition loss as large as 15 %**.

Thus, the catalysts of **Comparative Examples 2 and 3** are so weak that they are totally unsuitable for commercial use. Such catalysts should be removed from the discussion.

The remaining catalysts, i.e., the catalysts of **Comparative Examples 1, 4 and 5**, respectively, exhibit **yield values** (of the desired product) of 82.4 %, 81.9 % and 82.5 %, i.e., in the range of from 81.9 to 82.5 % (vs. 84.0 to 84.5 % in Examples 1 to 4).

The **yield values** (of the desired product) in the range of 84.0 to 84.5 % exhibited in **Examples 1 to 4** are very excellent values, as compared to the yield value range of from 81.9 to 82.5 % exhibited in **Comparative Examples 1, 4 and 5**. Even the **difference** between the **lower** limit value (84.0 %) of the yield value range exhibited in **Examples 1 to 4** and the **upper** limit value (82.5 %) of the yield value range exhibited in **Comparative Examples 1, 4 and 5** is as large as 1.5 %. Moreover, the **difference** between the **upper** limit value (84.5 %) of the yield value range exhibited in **Examples 1 to 4** and the **lower**

limit value (81.9 %) of the yield value range exhibited in **Comparative Examples 1, 4 and 5** is as large as **2.6 %**. In other words, **Examples 1 to 4** exhibit an improvement of **1.5 %** or **2.6 %** in the yield value, over the yield values exhibited in **Comparative Examples 1, 4 and 5**.

As described in item (I-2-1) above, even a small improvement (e.g., a 0.1 % improvement) in the **yield of the desired product** can achieve a **great commercial success** in the commercial production of (meth) acrylonitrile. Therefore, an improvement of **1.5 %** or **2.6 %** in the yield value achieved in **Examples 1 to 4**, over the yield values exhibited in **Comparative Examples 1, 4 and 5**, is a **very large improvement** which is commercially very advantageous.

As described hereinabove, the catalysts of **Examples 1 to 4** exhibit **both excellent yield of the desired product** and **excellent attrition resistance**, whereas the catalysts of **Comparative Examples 1 to 5** exhibit either a **poor yield of the desired product** or a **poor attrition resistance**.

Therefore, it is quite apparent that **only when** all requirements of **claim 1** of the present application are **satisfied**, there can be achieved the excellent effects of the present invention, i.e., the effects that the catalyst exhibits **not only a highly improved yield of the desired product, but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst.

As apparent from the above, the requirements of **claim 1** of the present application (especially the **pore distribution requirement**) are **critical** for achieving the excellent effects of the present invention (i.e., the effects that the catalyst exhibits **not only a highly improved yield of the desired product, but also a high attrition resistance** which renders

the catalyst suitable for commercial use as a fluidized-bed catalyst).

Such **criticality** of the requirements of **claim 1** of the present application (especially the **pore distribution requirement**) for achieving the above-mentioned excellent effects of the present invention, are quite unexpected from any prior art documents including the cited references.

**ITEMS 4-5: REJECTION OF CLAIMS 1 and 3-4 UNDER 35 U.S.C. § 102(b) AS BEING ANTICIPATED BY HINAKO et al. (JP2002-219362, hereafter D1).**

As recited in currently **amended independent claim1** and **new independent claims 8 and 10**, in applicant's invention, the reference of D1 does not anticipate the instant claims.

The Examiner stated that "D1 teaches a silica supported (ammoxidation) catalyst containing the metal oxides of molybdenum, vanadium, antimony, and niobium where the silica carrier is present in 40% by weight. See example 1, paragraph [0042] of D1. D1 teaches the catalyst with a particle diameter of from 5-200  $\mu\text{m}$  comprising at least 90% by weight of the catalyst. See Drawing 2 of D1 (there are no particles larger than 200  $\mu\text{m}$  or smaller than 5  $\mu\text{m}$ ). The catalyst of D1 also has pore size distribution wherein the pore volume of the pores having a pore diameter of 80  $\text{\AA}$  or less comprises less than 20% of the total pore volume and the pore volume corresponding to pores having a diameter of over 1000  $\text{\AA}$  is less than 20% of the total pore volume. See Drawing 1 of D1. Note, the preamble of claim 1 includes intended use, i.e. 'for use in producing acrylonitrile... fluidized bed reactor,' which does not impart patentable distinction to this composition. See MPEP 2112.02."

However, as shown in formula (1) of amended independent claim 1, three elements of Molybdenum, Bismuth and Iron are essential to the silica-carrying catalyst; as shown in formula (2) of new independent claim 8, five elements of Molybdenum, Bismuth, Cesium, Iron and Nickel are essential to the silica-carrying catalyst, and as shown in formula (3) of new independent claim 10, four elements of Molybdenum, Bismuth, Iron and Nickel are essential to the silica-carrying catalyst. No elements of **vanadium and Niobium** exist in formula (1)-(3) of current invention. On the other hand, D1 discloses that a silica-carrying catalyst contains Molybdenum, **Vanadium, Antimony, and Niobium** as constituent elements. Therefore, the silica-carrying catalysts of current invention are different from the one in D1.

In addition, another reason that the catalysts of current invention are different from the one in D1 is simply because of their different purposes. While the current invention is to obtain high activity in producing the desired product as well as high attrition resistance when used on a commercial scale (see page 9 of current specification), the purpose of D1 is to provide a high-flow silica-carrying catalyst (see Abstract of D1).

Furthermore, because of the different catalysts and different proposes between current invention and D1, they showed remarkable difference in their features (see Table 2 of current specification and Table 1 of D1). The catalysts of current invention provide higher propane conversion percent and higher Acrylonitrile selectivity percent than the one of D1. Also, the catalysts of current invention provide high Attrition resistance which has not been disclosed by D1.

**The Applicants want to add further response as follows.**

(The English of the computer translation of D1, supplied from the U.S. Patent Office, is poor. Therefore, the Applicants hereby submit **Exhibit 1**, which is a sworn **partial English translation of D1**. A copy of D1 (JP-A-2002-219362) was submitted as an IDS reference on October 4, 2005.)

As described below, contrary to the Examiner's statement, the particulate porous ammoxidation catalyst of the instantly amended claim 1 has **novelty and non-obviousness** over the catalyst of D1.

The silica-supported catalyst of D1 contains metal oxides of molybdenum, vanadium, antimony and niobium. That is, molybdenum (Mo), **vanadium (V)**, antimony (Sb) and **niobium (Nb)** are **essential** in the catalyst of D1. The essentiality of these elements in the catalyst of D1 is apparent from, for example, claim 1 of D1. For easier reference, claim 1 of D1 is quoted below:

"[Claim 1] A silica-supported catalyst for use in gaseous phase catalytic oxidation or ammoxidation of propane or isobutane for producing an unsaturated carboxylic acid or an unsaturated nitrile,

said catalyst satisfying the following requirements (a), (b) and (c):

(a) said catalyst comprises a composition represented by the following formula (I):



wherein:

Z is at least one element selected from the group consisting of W, Cr, Ti, Al, Ta, Zr, Hf, Mn, Re, Fe, Ru, Co, Rh, Ni, Pd, Pt, Cu, Ag, Zn, B, Ga, In, Ge, Sn, P, Pb, Bi, Y, rare earth elements and alkaline earth metals; and

a, b, c, d, and n are, respectively, the atomic ratios of vanadium (V), antimony (Sb), niobium

(Nb), Z and oxygen (O), relative to molybdenum (Mo),

wherein:

**0.1 ≤ a ≤ 1,**

**0.01 ≤ b ≤ 0.6,**

**0.01 ≤ c ≤ 0.3,**

**0 ≤ d ≤ 1, and**

n is a number determined by and consistent with the oxidation states of the metal elements present,

(b) the oxide represented by the formula (I) is supported on a silica carrier, said silica carrier being present in an amount of from 20 to 60 % by weight, and

(c) said catalyst has a pore volume of 0.15 cm<sup>3</sup>/g or more."

*(see claim 1 of D1; see pages 1 to 3, item (2) of Exhibit 1, which is a partial English translation of D1)*

Further, all catalysts produced in the Working Examples of D1 contain the above-mentioned essential metal elements, i.e., molybdenum (Mo), **vanadium (V)**, antimony (Sb) and **niobium (Nb)** (see Table 1 at page 11 of D1; see pages 3 and 4, item (3) of Exhibit 1).

On the other hand, as seen in the claim Amendment, in order to more clearly define the present invention, **vanadium (V)** and **niobium (Nb)** have been **deleted** from the group of candidate component elements for the optional metal element D of the metal oxide of the formula (1) which has been inserted in claim 1.

Also, claim 4 (directed to a method for producing the catalyst of claim 1) has been amended in accordance with the amendment to claim 1.

Further, any of all catalysts produced in the Working Examples of the present

specification does not contain any of **vanadium (V)** and **niobium (Nb)** (see page 72, Table 1 of the present specification).

Therefore, the instantly amended claim 1 of the present application has **novelty** over the catalyst of D1.

Further, it should be noted that the catalyst of claim 1 of the present application has also **non-obviousness** over the catalyst of D1. This point is described below.

As described in item (I-3) above in detail, the requirements of **claim 1** of the present application (especially the pore distribution requirement) are **critical** for achieving the excellent effects of the present invention (i.e., the effects that the catalyst exhibits **not only** a **highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst).

Such **criticality** of the essential features of claim 1 of the present application for achieving such **excellent effects** of the present invention is quite **unexpected** from D1.

In addition, it should be noted that, in D1, improvement of the **yield** of the desired product is **not** important. Also, in D1, improvement of the **attrition resistance** of a catalyst is **not** important. This is apparent from the below-described **points (1), (2) and (3)**.

As the point (1), the following should be noted. D1 has the following description:

"[0066]

[Effect of the invention] The silica-supported catalyst of the present invention exhibits a large volume while maintaining the reaction results at the same level as in the case of the conventional silica-supported catalysts. Therefore, the catalyst exhibits a high flowability and hence a high efficiency of gas-catalyst contact, thereby facilitating

the operation of a fluidized bed reactor."

*(emphasis added) (see paragraph [0066] of D1; see page 5, item (4) of Exhibit 1, which is a partial English translation of D1)*

As apparent from the above-quoted description of D1, D1 recognizes that the invention of D1 cannot improve the reaction results (such as the **yield** of the desired product). In addition, improvement of the **attrition resistance** of the catalyst is not mentioned as an effect of the invention of D1. The invention of D1 is aimed at improving a **catalyst flowability**.

As the point (2), the following should be noted. In agreement with the point 1 above, the reaction results (in terms of propane **conversion** and acrylonitrile **selectivity**) (i.e., reaction productivity) achieved in the **Working Examples** of D1 largely overlaps with the reaction results achieved in **Comparative Working Examples** of D1 (see Table 1 at page 11 of D1; see pages 3 and 4, item (3) of Exhibit 1). For easier reference, the data of **propane conversion** and **acrylonitrile selectivity**, as shown in Table 1 at page 11 of D1 (see pages 3 and 4, item (3) of Exhibit 1) is quoted below.

More specifically, the propane **conversion value range** (from 48.0 to 50.1) and acrylonitrile **selectivity value range** (from 63.0 to 67.8) achieved in the **Working Examples** of D1, respectively, largely overlap with the propane **conversion value range** (from 48.2 to 48.6) and acrylonitrile **selectivity value range** (from 63.2 to 67.8) achieved in **Comparative Working Examples** of D1 (see Table 1 at page 11 of D1; see pages 3 and 4, item (3) of Exhibit 1). (Needless to say, since an acrylonitrile **yield** is obtained by the formula: **(propane conversion) × (acrylonitrile selectivity)**, the above-mentioned "reaction results"

or "reaction productivity" can be reworded as "acrylonitrile yield".)

As the point (3), it should be noted that, as seen from Table 1 of D1, the **attrition resistance** of a catalyst is not evaluated in the Working Examples and Comparative Working Examples of D1. This shows that D1 pays **no** particular attention to improvement of **attrition resistance**.

Thus, the points (1), (2) and (3) above clearly show that, in D1, improvement of the **yield** of the desired product is not important, and improvement of the **attrition resistance** of a catalyst is not important.

Therefore, it is apparent that the disclosure of D1 cannot give any teaching or suggestion which is useful for improving any of the **yield** of the desired product and the **attrition resistance** of a catalyst. Hence, the present invention cannot be easily reached at from the disclosure of D1.

Further, in the method of the instantly amended claim 4 for producing the catalyst of claim 1, the component metal elements employed have been limited to the metal elements of the metal oxide represented by the formula (1) of the instantly amended claim 1. Therefore, the method of claim 4 can not be taught or suggested by D1 taken in any respect.

As apparent from the above, claims 1, 3 and 4 of the present application have **novelty** and **non-obviousness** over the catalyst of D1. D1 does not teach or suggest the essential features of the present invention or the effects thereof.

It is firmly believed that the rejection of claims 1, 3 and 4 over D1 under 35 U.S.C. 102 has been removed by the above argument and data.

Accordingly, Applicants respectfully submit that an anticipation rejection cannot be

based on Hinako et al. patent publication and allowance of the pending independent claim 1 as amended, new independent claims 8 and 10 and depending claims 3-7, 9, and 11 are respectfully requested.

**ITEMS 6-8: REJECTION OF CLAIMS 2 and 6-7 UNDER 35 U.S.C. § 103(a) AS BEING UNPATENTABLE OVER D1, AS APPLIED TO CLAIM 1 ABOVE, IN VIEW OF UMEMURA et al. (US4264476, hereafter D2).**

In items 6-8 of the office action, claims 2 and 6-7 have been rejected under 35 U.S.C. 103(a) as being unpatentable over D1, as applied to claim 1 above, in view of Umemura et al.. More specifically, the Examiner states as follows:

“D1 teaches the elements of claim 1. See 102 rejection above. However, D1 does not teach the composition as required by claim 2. D1 does not disclose using iron and bismuth in composition as required by claim 2, but does not teach the presence of an alkali metal in the composition in the given proportions required by claim 2.

.....  
.....  
.....  
.....

Though D1 does not teach the presence of sodium, potassium, rubidium, or cesium; it would have been obvious to one of ordinary skill to add one these metals as specified in D2. ....”

**The Applicants want to respond as follows.**

The catalyst of the instantly amended independent claim 1 and new independent claims 8 and 14 of the present application (which does not contain any of vanadium and niobium) can not be easily made from any combination of D1 and D2.

Further, the **criticality** of the essential features of claim 1, 8 and 10 of the present application for achieving the **excellent effects** of the present invention (i.e., the effects that the catalyst exhibits **not only a highly improved yield** of the desired product, **but also a high attrition resistance** which renders the catalyst suitable for commercial use as a fluidized-bed catalyst) is quite **unexpected** from any combination of D1 and D2.

Thus, the catalyst of the instantly amended claim 1 and new claims 8 and 10 of the present application have **non-obviousness** over any combination of D1 and D2. D1 and D2 do not teach or suggest the essential features of the present invention or the effects thereof.

Accordingly, even combining the teaching of the Hinako et al patent publication with the teaching of the Umemura et al patent, any one of ordinary skill in this art would not be motivated at all. Therefore, applicants respectfully submit that an obviousness rejection cannot be based on Hinako et al. in view of Umemura et al and allowance of the pending independent claim 1 as amended, new independent claims 8 and 10 and depending claims 3-7, 9, and 11 are respectfully requested.

**ITEM 9: REJECTION OF CLAIM 5 UNDER 35 U.S.C. § 103(a) AS BEING UNPATENTABLE OVER D1 IN VIEW OF SASAKI et al. (US4370279, hereafter D3).**

**The Applicants want to respond as follows.**

Claim 5 depends from claim 4 (directed to a method for producing the catalyst of claim 1).

It is believed that the rejection of claim 4 has been removed by the instant amendment to claim 4. Therefore, it is also believed that the rejection of claim 5 has been removed.

Accordingly, even combining the teaching of the Hinako et al patent publication with the teaching of the Sasaki et al patent, any one of ordinary skill in this art would not be motivated at all. Therefore, applicants respectfully submit that an obviousness rejection cannot be based on Hinako et al. in view of Sasaki et al and allowance of the pending independent claim 1 as amended, new independent claims 8 and 10 and depending claims 3-7, 9, and 11 are respectfully requested.

Table 1 of D1

Composition of catalyst					Catalyst preparation conditions			Physical properties of catalyst			Ammoxidation of propane		
Mo	V	Sb	Nb	SiO <sub>2</sub> (wt%)	.....	.....	Pore volume (cm <sup>3</sup> /g)	Bulk density (g/cm <sup>3</sup> )	spherical index	contact time (S)	propane conversion (%)	acrylonitrile selectivity (%)	
Example 1	1	0.3	0.23	0.07	....	....	....	....	....	...	48.6	63.8	
Example 2	1	0.3	0.23	0.07	....	....	....	....	....	...	50.1	63.0	
Comparative Example 1	1	0.3	0.23	0.07	....	....	....	....	....	...	48.5	63.9	
Comparative Example 2	1	0.3	0.23	0.07	....	....	....	....	....	...	48.6	63.9	
Example 3	1	0.23	0.25	0.09	....	....	....	....	....	...	48.0	67.8	
Example 4	1	0.23	0.25	0.09	....	....	....	....	....	...	49.3	67.7	
Comparative Example 3	1	0.23	0.25	0.09	....	....	....	....	....	...	48.2	67.8	
Example 5	1	0.3	0.22	0.07	....	....	....	....	....	...	48.0	63.2	
Comparative Example 4	1	0.3	0.22	0.07	....	....	....	....	....	...	48.3	63.2	

## CONCLUSION

Thus, it is believed that all rejections and objections have been removed, and the present application is now in condition for allowance.

Reconsideration and early favorable action on the claims are earnestly solicited.

There being no further outstanding objections or rejections, it is submitted that the application is in condition for allowance. An early action to that effect is courteously solicited.

Finally, if there are any formal matters remaining after this response, the Examiner is requested to telephone the undersigned to attend to these matters.

If there are any additional fees associated with filing of this Amendment, please charge the same to our Deposit Account No. 19-3935.

Respectfully submitted,

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